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Turbulent Schmidt number from a tracer experiment

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Abstract

Measurements of pesticide emission from a bare soil were used to calculate the turbulent Schmidt number (*Sc*): the ratio of eddy diffusivity for momentum (eddy viscosity) to the diffusivity for tracer mass. The value of *Sc* has implications for the measurement of trace gas emissions, and there is a broad range of reported values for the atmospheric surface layer. During our experiment *Sc* averaged 0.6, with large variability between observation periods. The standard deviation in *Sc* was 0.31, with no obvious correlation to atmospheric conditions. Some of this variability is due to measurement uncertainty, but we believe it also reflects true variability in *Sc*. We show that flux-gradient formula, which assume higher values of *Sc*, underestimate the true tracer emission rate *Q*. We also show that a dispersion model with $Sc = 0.6$, does better at inferring *Q* than a model with $Sc = 0.45$. Published by Elsevier Science B.V.

Keywords: Schmidt number; Tracer emmisions; Eddy viscosity; Flux-gradient method; Lagrangian stochastic models; Atmospheric dispersion; Diffusivity

1. Introduction

First-order closure (*K*-theory) is a common approximation in atmospheric transport problems. Conventional *K*-theory formulae for the time-average vertical fluxes of tracer mass (F_c) and momentum (F_m) are:

$$F_c = -K_c \frac{\partial C}{\partial z}, \qquad F_m = \rho K_m \frac{\partial S}{\partial z} \quad (= \rho u_*^2), \tag{1}$$

where K_c and K_m are tracer and eddy diffusivities, C the average tracer concentration, S the average wind speed, ρ the air density, and z the vertical dimension (u_* is the friction velocity). Although, K_m in the atmospheric surface layer is reasonably well known, this is not so for K_c. This uncertainty can be conveniently expressed as uncertainty in the turbulent Schmidt number (*Sc*):

$$Sc = \frac{K_m}{K_c}. \tag{2}$$

The near-surface value of *Sc* is the subject of this paper, and we focus on two examples where knowledge of *Sc* is important for the measurement of tracer emissions from the surface.

The flux-gradient (FG) technique is often used to estimate emissions where it is assumed that the emission rate Q equals F_c in Eq. (1). In the aerodynamic FG method, Eq. (1) are combined and Q is calculated from measurements of C and S near the ground:

$$\frac{Q}{u_*^2} = -\frac{K_c\ \Delta C}{K_m\ \Delta S} = -\frac{\Delta C}{Sc\ \Delta S}, \tag{3}$$

where height differences ΔC and ΔS have been substituted for the gradients. The value of *Sc* is therefore crucial to FG measurements (Eq. (3) is usually

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modified to give working formulae where Sc may not appear explicitly, but a value is presumed).

Dispersion models are also useful tools for estimating emissions. By predicting the relationship between Q and downwind concentration, they allow a measurement of C to establish Q (e.g. Wilson et al., 1983). But such predictions are sensitive to the effective Sc of the model. Consider Lagrangian stochastic (LS) dispersion models. Sawford and Guest (1988) derived an expression for the far-field K_c of models formulated according to criteria given by Thomson (1987), and using the standard Monin–Obukhov expression for K_m, we can write a model Sc as:

$$Sc = \frac{K_m}{K_c} = \frac{(ku_* z/\phi_m)}{2(\sigma_w^4 + u_*^4)/(C_0 \epsilon)} = \frac{ku_* z \epsilon}{2(\sigma_w^4 + u_*^4)\phi_m} C_0, \tag{4}$$

where k is the von Karman constant, ϕ_m the nondimensional wind shear, σ_w the standard deviation of the vertical velocity fluctuations, ϵ the turbulent kinetic energy dissipation rate (often presumed proportional to u_*^3), and C_0 is a 'universal' turbulent constant (reported values from 3 to 9). Parameterizations of σ_w, ϵ, ϕ_m and the chosen value of C_0 determine the model Sc.

A large range of boundary layer Sc values is found in the meteorological and engineering literature. The 'classic' micrometeorological assumption is equality between the eddy diffusivities for momentum, heat, moisture, and mass, so that Sc is equal to the Prandtl number (Pr)[1] and equal to one (Oke, 1978; Garratt, 1992). Examples of this are found in the FG measurements of Denmead et al. (1978) and Nemitz et al. (2000), and the dispersion models of Legg (1983) and Ley (1982). Another assumption is $Sc = 0.75$ (Stull, 1988). This is corroborated by observations of Pr and Sc in the wind tunnel (Daily and Harleman, 1966; Launder, 1978; Batt et al., 1999), and the success of dispersion models with $Sc = 0.75$ (Nieuwstadt and van Ulden, 1978; Gryning et al., 1983). Yet another common assumption is that $Sc = 0.6$. This value is supported by the pipe flow measurements of Hinze (1975), and the successful dispersion models of Wilson et al. (1981) and Reid (1979). Some of the variability in reported Sc may be due to a height dependence in the boundary layer. Raupach et al. (1996) argue that Pr changes dramatically with height within and above a plant canopy. And in a wind tunnel simulation, Koeltzsch (2000) show a strong height gradient in Sc, with a peak near one-third the boundary layer depth.

In this study, a tracer dispersion experiment provided an opportunity to examine the value of Sc near the ground. The original motivation for our experiment was to take FG measurements of pesticide volatilization from a bare field. But including an independent estimate of tracer flux (the integrated horizontal flux technique) allowed us to calculate Sc and examine the implication for emission measurements.

2. Tracer experiment

The pesticide metolachlor was applied to a large bare field in Iowa (Fig. 1). After application, the average metolachlor concentration (C) and wind speed (S) were measured near the center of the field (the 'FG tower') at heights $z = 0.15, 0.29, 0.55, 1.05$, and 2 m above ground. These observations were later used to calculate FG estimates of pesticide emission rates (Q_{FG}). The minimum fetch (distance to the upwind edge of the field) at the FG tower was 140 m. The concentration C was found by drawing air through foam plugs that absorbed metolachlor. The sampling period lasted either 2 or 4 h. The metolachlor mass was determined by a GC/MS analysis (described in more detail in Prueger et al., 1999). Cup anemometers were used to measure S. The experiment lasted 5 days, and we measured 62 C and S profiles.

2.1. The IHF technique

The integrated horizontal flux (IHF) technique provided our measurement of metolachlor emissions (Q_{IHF}). The IHF is a simplified mass-balance method to estimate the horizontal flux of a tracer past a tower (Denmead et al., 1977). An IHF tower was placed 15 m from either the north or south edge of the field, depending on wind direction (Fig. 1). Metolachlor concentration was measured at the same heights as

[1] Pr is the ratio of eddy diffusivity for momentum to the diffusivity for heat. Monin and Yaglom (1965) argue that mass and heat transfer occur by a similar mechanism, and $Sc = Pr$ in moderate stratification.

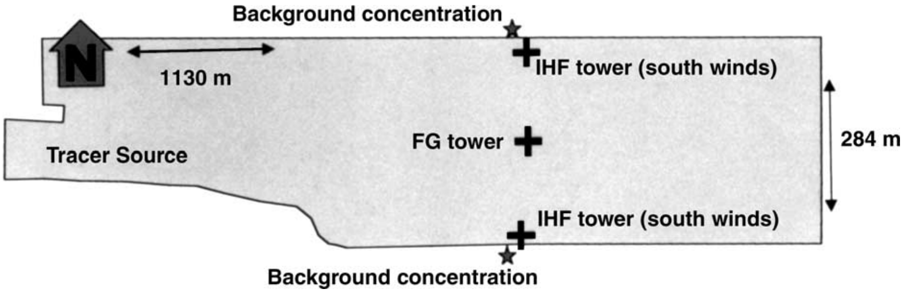

Fig. 1. Layout of tracer experiment. The tracer source was a large bare field sprayed with metolachlor. Metolachlor concentration profiles were measured at two towers, a flux-gradient (FG) tower near the center of the field, and an integrated horizontal flux (IHF) tower at either the north or south edge of the field (depending on wind direction). Background concentration was measured upwind of the field.

the FG tower. The emission rate was calculated as:

$$Q_{\mathrm{IHF}} = \frac{\cos\theta}{X}\sum_{i=1}^{5} S_i C_i \Delta z_i \quad \left(\approx \frac{1}{X}\int_0^{\infty} \langle uc \rangle \, \mathrm{d}z\right), \tag{5}$$

where X is the distance from the tower to the field edge (15 m), θ the average wind angle from normal to the field edge, i the index for observation height, and Δz_i the depth of the layer attributed to each measurement height. An upwind (background) concentration of metolachlor was measured at each observation time, and found to be insignificant.

Advantages of the IHF method are simplicity and lack of assumptions (i.e. it is model-free). However, there are two concerns. The first is the neglect of turbulent horizontal flux. Eq. (5) only approximates the true flux given by the height integral of $\langle uc \rangle$, the time-average product of the wind velocity normal to the field edge and concentration. The result is that Q_{IHF} overestimates the true flux, reportedly by between 5 and 20% (Denmead and Raupach, 1993). We compensated for this error by reducing Q_{IHF} by 20%. This correction factor was chosen after simulating the geometry of our problem with a 3D LS model. We used standard parameterizations of the surface layer wind flow, accounting for the effect of stability and wind direction, to estimate the magnitude of the horizontal turbulent flux.

A second concern with IHF was the possibility of unaccounted tracer flux above our top C measurement at $z = 2$ m. This will certainly occur if the wind angle θ is far from normal to the field edge (i.e. large fetch). We sought to avoid this by rejecting periods when the upwind distance to the field edge ($X/\cos\theta$) was greater than 25 m, as well as those periods bracketing a change in wind direction from northerly to southerly, and vice versa. With this constraint, we accepted 19 Q_{IHF} observation periods. As partial confirmation that we did not overlook flux, we looked at an alternative to Eq. (5). We fit profiles to the observations of S and C (using a log function which asymptotes to zero at large heights), and integrated their product to $z = 5$ m. If the C observations indicate significant tracer mass above our observation height, then this calculation should differ from the simple summation in Eq. (5). However, the two calculations did not systematically differ (average difference being 1%).

Even with an accurate correction for turbulent flux, and a careful choice of observation periods, there is uncertainty in Q_{IHF} due to random measurement errors in S and C. We made two conservative assumptions: that fractional uncertainty in our S measurements ($\delta S/S$) was 5%, and in C ($\delta C/C$) was 20% (Taylor, 1982). An uncertainty in Q_{IHF} is calculated as:

$$\delta Q_{\mathrm{IHF}} = \frac{\cos\theta}{X}\sqrt{\sum_{i=1}^{5}\left(\left(\frac{\delta S}{S}\right)^2 + \left(\frac{\delta C}{C}\right)^2\right) S_i^2 C_i^2 \Delta z_i^2}. \tag{6}$$

An analysis of our data shows that the resulting uncertainty in Q_{IHF} is approximately 10%.

2.2. Determining u_*

In our analysis, u_* and L were determined using a profile approach. A best-fit u_* and L were chosen to give agreement between our measurements of S and sensible heat flux (CSAT-3 sonic anemometer, Campbell Sci.), and a standard Monin–Obukhov wind profile (Hogstrom, 1996). A best-fit z_0 of 0.008 m was used. This profile approach for getting u_* was chosen over a velocity covariance measurement from the sonic anemometer (i.e. $u_* = (\langle u'w'\rangle^2 + \langle v'w'\rangle^2)^{1/4}$). The u_* from the profile-fitting ($u_{*\mathrm{p}}$) and the sonic anemometer ($u_{*\mathrm{s}}$) did not agree, with $u_{*\mathrm{s}} \sim 0.8$ ($u_{*\mathrm{p}}$). This is an unexplained aspect of the study. We chose to accept ($u_{*\mathrm{p}}$) for the following reasons: confidence in our S measurements; inability to post-rotate the sonic statistics to correct for leveling (we did not take enough information); the sensitivity of the covariances to flow distortion from the sonic housing; the need to maintain consistency with FG and IHF observations which used the S profile; and that ($u_{*\mathrm{p}}$) should ideally be a more spatially representative indication of momentum flux than the point measurement ($u_{*\mathrm{s}}$). Fortunately, the results of this study are not sensitive to the consistent use of either ($u_{*\mathrm{p}}$) or ($u_{*\mathrm{s}}$): Sc is not significantly different whether we use ($u_{*\mathrm{p}}$) and accept the cup anemometer S profile, or use ($u_{*\mathrm{s}}$) and generate a sonic consistent S profile as a replacement for the cup profile (the difference Sc was 3%).

3. The turbulent Schmidt number

3.1. Calculation

The turbulent Sc was calculated using the standard Monin–Obukhov definition of K_m, and accepting Q_IHF as the true tracer flux:

$$Sc = \frac{K_\mathrm{m}}{K_\mathrm{c}} = -\frac{(ku_* z_\mathrm{g}/\phi_\mathrm{m})}{Q_\mathrm{IHF}\,\Delta z/\Delta C}, \tag{7}$$

where ΔC is taken from the central FG tower, and z_g the geometric mean of the observation heights for C (e.g. $z_\mathrm{g} = (z_1 z_2)^{0.5}$). We have assumed $k = 0.4$, and used the ϕ_m relationships suggested by Hogstrom (1996):

$$\begin{aligned} \phi_\mathrm{m} &= \left(1 - 19\frac{z}{L}\right)^{-0.25} \quad (L < 0),\\ \phi_\mathrm{m} &= 1 + 5.3\frac{z}{L} \quad (L > 0). \end{aligned} \tag{8}$$

Because our C profile consists of five observation heights, we can calculate 10 Sc values at each observation period by using all possible height combinations in Eq. (7). We initially grouped values by height, but there was no evidence of height dependence. We thereafter grouped all data together, giving an average Sc for each of the 19 observation periods.

3.2. Error estimates

A fractional uncertainty in Sc calculated from Eq. (7) can be expressed as a function of the fractional uncertainties in u_*, Q_IHF, ΔC, and ϕ_m ($\delta u_*/u$, etc.):

$$\frac{\delta Sc}{Sc} = \sqrt{\left(\frac{\delta u_*}{u_*}\right)^2 + \left(\frac{\delta \Delta C}{\Delta C}\right)^2 + \left(\frac{\delta \phi_\mathrm{m}}{\phi_\mathrm{m}}\right)^2 + \left(\frac{\delta Q_\mathrm{IHF}}{Q_\mathrm{IHF}}\right)^2}, \tag{9}$$

assuming k and z_g are known precisely (see Taylor, 1982). We assume an uncertainty in u_* and ϕ_m of 5 and 10%, respectively. Our earlier calculation gave an uncertainty in Q_IHF of 10% (Section 2.1). We calculate the uncertainty in ΔC using our actual C data, assuming an uncertainty in C of 20%. For two observations C_1 and C_2:

$$\frac{\delta \Delta C}{\Delta C} = \frac{\sqrt{(\delta C_1)^2 + (\delta C_2)^2}}{|C_1 - C_2|}, \tag{10}$$

and we found $\delta\Delta C/\Delta C$ averaged 61%. These individual uncertainties result in an uncertainty in a single Sc observation of 63%. However, for each observation period we calculate an average Sc from 10 values (i.e. different height combinations), and the uncertainty of this average is reduced by the reciprocal of the square root of the number of samples. Therefore, we estimate that our average Sc observations have an uncertainty of 20%.

3.3. Value of Sc

During our experiment the average Sc was 0.60, meaning the vertical diffusivity of tracer mass was greater than the diffusivity of momentum (or eddy viscosity). This is less than the classic micrometeorological assumption of one, but in agreement with other studies (Hinze, 1975; Wilson et al., 1981). The dominant feature of our observations was large Sc varia-

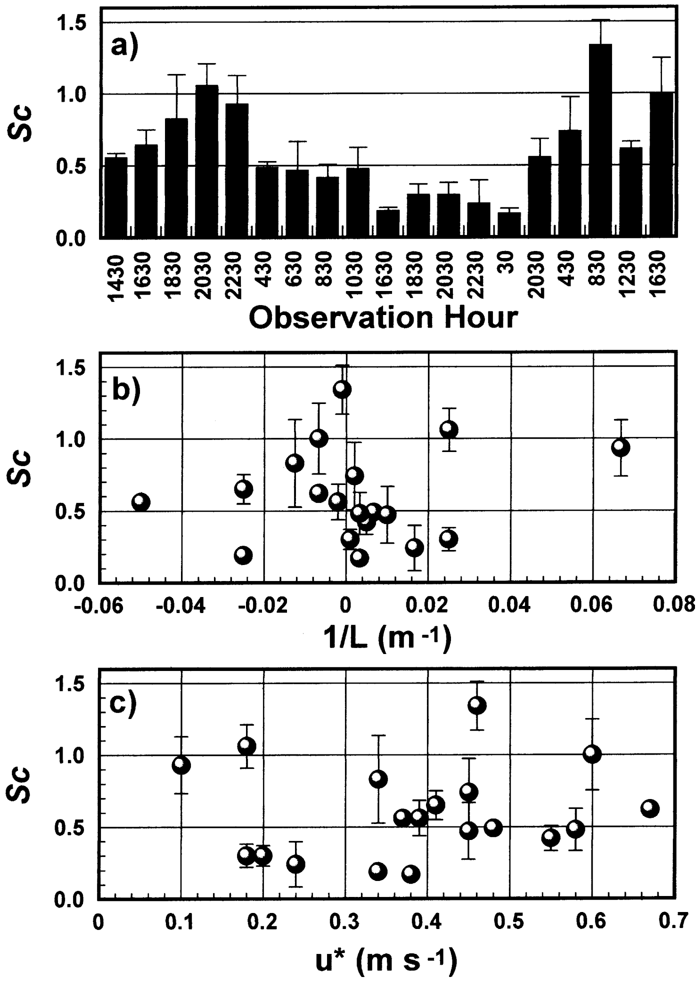

Fig. 2. The turbulent Schmidt number *Sc* vs.: (a) observation time (observations are in chronological order, but are not continuous in time); (b) the reciprocal of the Obukhov length *L*, and (c) the friction velocity u_*. Error bars give the standard error of the *Sc* observations.

bility. Over the 19 observation periods, the standard deviation of *Sc* was 0.31 (see Fig. 2), with values ranging from 0.17 to 1.34. Some variability can be attributed to measurement error, which we estimate to be 20%.[2]

We expected a relationship between *Sc* and atmospheric stability. Hogstrom's (1996) analysis of atmospheric data indicates that *Pr* increases as stability changes from unstable to stable, and we might expect the same for *Sc*. However, there was no clearly identified trend in *Sc* with either stability, time of day, or wind speed (Fig. 2). We see only the slightest indication of a stability effect when we segregate our observations: $Sc = 0.56$ in unstable conditions ($-100 < L < 0$), 0.60 in the near-neutral cases ($|L| \geq 100$), and 0.63 in stable conditions ($0 < L < 100$). But there is no statistical difference in *Sc* between these categories.

Some of the extremes in *Sc* may be due to inappropriate averaging periods. The two largest observations (1.34 and 1.06) spanned a sunrise and sunset (stability trended from moderately stable to moderately

[2] Our calculated uncertainty for an individual *Sc* observation was 63%, which agreed well with the measured within-period standard deviation of 68%. This suggests our assumptions of measurement uncertainty are reasonable.

unstable, and vice versa). In such a rapidly changing environment, the association of long-interval average C and S profiles (and the derived L and u_*) to an average vertical flux may not be reasonable. At the other extreme there was a succession of low Sc values during the middle of the experiment, with five observations below 0.35. Yet nothing was obviously different with this period, either in terms of wind speed, stability, wind direction, or time of day.

It is possible that some Sc variability may be due to the false assumption of flux homogeneity across the landscape. The IHF tower is far removed from the FG tower, and their 'footprints' (ground area influencing measurements) are quite different. Spatial variation in Q will mean Q_{IHF} and ΔC in Eq. (7) are no longer linked to the same vertical flux. Could there be a 25–50% variability in Q across our field, over averaging scales of order 15 m (the scale of the measurement footprints)? This level of variability in sprayer application rates seems unlikely, and the homogeneity of the site would seem to argue against such a large landscape induced variability.

It seems more likely that the variability we observed in Sc is a real feature of the atmosphere, due to the shortcomings of a Monin–Obukhov representation of the surface layer and a K-theory description of atmospheric transport. This is disconcerting for both the measurement of surface fluxes and dispersion modeling, as it suggests significant uncertainty is a characteristic of these calculations.

4. Implications for flux measurement

Consider a commonly used FG formula (Oke, 1978) where u_* in Eq. (3) is replaced with the standard Monin–Obukhov flux-gradient definition:

$$Q_{\mathrm{FG}} = -\frac{k^2}{\phi_{\mathrm{m}}\phi_{\mathrm{c}}}\frac{z_{\mathrm{g}}^2}{\Delta z^2}\Delta C \Delta S, \qquad (11)$$

where z_{g} is the geometric height of the measurement levels, and ϕ_{m} and ϕ_{c} the universal stability corrections for momentum and mass flux. The choice of ϕ_{m} and ϕ_{c} implies Sc (i.e. $Sc = \phi_{\mathrm{c}}/\phi_{\mathrm{m}}$). We examine three potential choices:

- Hg: $\phi_{\mathrm{c}} = \phi_{\mathrm{m}}$ (i.e. $Sc = 1$): ϕ_{m} formula from Hogstrom (1996) given in Eq. (7).
- DB: $\phi_{\mathrm{c}} = \phi_{\mathrm{h}}$ (i.e. $Sc = Pr$): ϕ_{m} and ϕ_{h} formulae from Dyer (1974) and Dyer and Bradley (1982):

$$\begin{aligned} &\phi_{\mathrm{m}} = \left(1 - 15\frac{z}{L}\right)^{-1/4}, \\ &\phi_{\mathrm{c}} = \phi_{\mathrm{h}} = 0.95\left(1 - 14\frac{z}{L}\right)^{-1/2}, \quad (L < 0) \\ &\phi_{\mathrm{m}} = 1 + 4.8\frac{z}{L}, \\ &\phi_{\mathrm{c}} = \phi_{\mathrm{h}} = 0.95 + 4.5\frac{z}{L}, \quad (L > 0) \end{aligned} \qquad (12)$$

- PML: ϕ_{m} and ϕ_{c} formula suggested by Pruitt et al. (1973):

$$\begin{aligned} &\phi_{\mathrm{m}} = \left(1 - 16\frac{z}{L}\right)^{-1/3}, \\ &\phi_{\mathrm{c}} = 0.89\left(1 - 22\frac{z}{L}\right)^{-0.4}, \quad (L < 0) \\ &\phi_{\mathrm{m}} = \left(1 + 16\frac{z}{L}\right)^{1/3}, \\ &\phi_{\mathrm{c}} = 0.89\left(1 + 34\frac{z}{L}\right)^{0.4}, \quad (L > 0) \end{aligned} \qquad (13)$$

Each choice implies a larger Sc than we observed, with DB and PML formulae giving an Sc which varies with height and stability (see Fig. 3). The result of a too-large Sc is underpredicted emissions. This can be

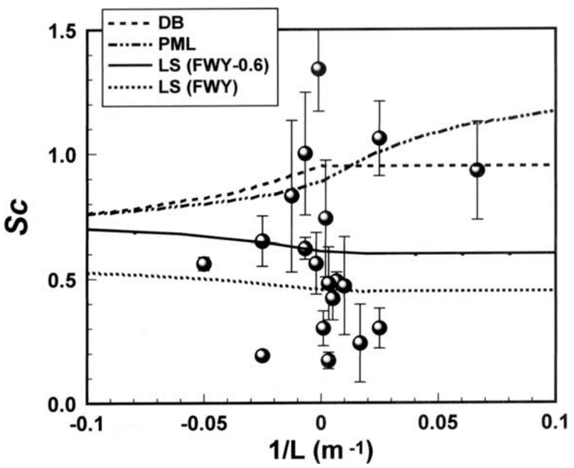

Fig. 3. Observed Schmidt number Sc (symbols) vs. the reciprocal of the Obukhov length L. Error bars give the standard error of the Sc observations. The lines represent values of Sc: (1) implied by FG formula given in the text (DB, and PML), (2) from the LS dispersion model of Flesch et al. (1995) (FWY), and (3) from the Flesch et al. model modified to give $Sc = 0.60$ at neutral stability (FWY-0.6). Values for $Z = 1$ m.

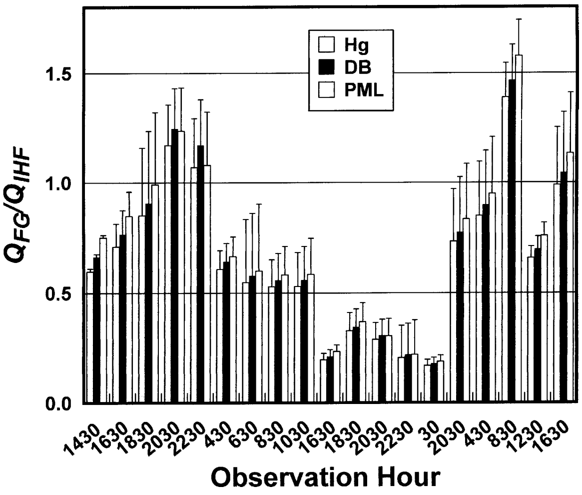

Fig. 4. Ratio of FG estimates of emission rate (Q_{FG}) to the IHF estimates (Q_{IHF}) for different observation times. Three FG formulae are shown (Hg, DB, and PML). Error bars are the standard error of the FG calculations from 10 height combinations.

seen when using these formula with our C and S observations to give Q_{FG} (Fig. 4). The average $Q_{\mathrm{FG}}/Q_{\mathrm{IHF}}$ was 0.65 for Hg, 0.70 for DB, and 0.73 for the PML formula. If Q_{IHF} is correct, these FG formula underestimate tracer emissions by about 30%. And if the large variability we saw in Sc is also real, then these FG estimates will have a similarly large uncertainty. Our data suggests a simpler FG formula:

$$Q_{\mathrm{FG}} = -\frac{k^2}{Sc\phi_{\mathrm{m}}^2}\frac{z_{\mathrm{g}}^2}{\Delta z^2}\Delta C \Delta S, \tag{14}$$

where $Sc = 0.6$. Given a large random variability in Sc, there would be little reason to use a more complex relationship.

When dispersion models are used to infer emission rates, their results will be sensitive to the model Sc. Flesch et al. (1995) used an LS model to infer emission rates (Q_{LS}) from an observed C. Their parameterizations resulted in a model Sc of 0.45 in neutral conditions (see Fig. 3). Therefore, this model is over-diffusive, and in our case, should give erroneously high Q_{LS}. Indeed, using the Flesch et al. (1995) model with our C observations at the FG tower gives Q_{LS} averaging 23% higher than Q_{IHF} (see Fig. 5). But if we change the model in a simple way to give $Sc = 0.6$ in neutral conditions, by multiplying the Lagrangian timescale by 0.75 (see Fig. 3), then

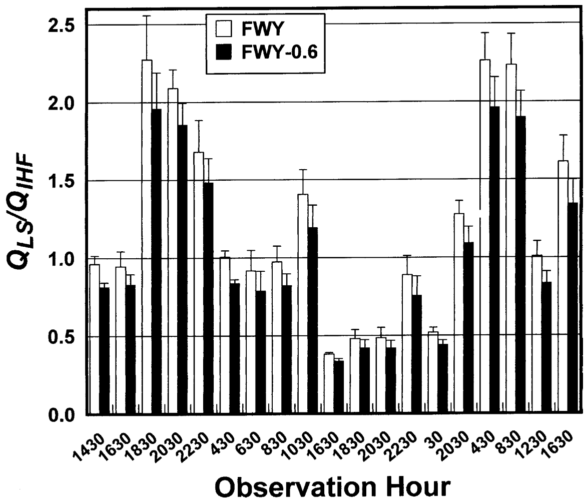

Fig. 5. Ratio of the LS estimates of emission rate (Q_{LS}) to the IHF estimates (Q_{IHF}) for different observation times. The two LS predictions are from the Flesch et al. (1995) model (FWY), and the FWY model modified to give $Sc = 0.60$ in neutral stability (FWY-0.6). Error bars are the standard error of the LS predictions from the five C observation heights.

Q_{LS} averaged only 6% higher than Q_{IHF} (Fig. 5). A dispersion model must get more than just Sc correct to predict Q (e.g. average wind profile and velocity variances), but in this case, a more accurate representation of Sc leads to a more accurately predicted Q_{LS}.[3]

5. Conclusions

Our tracer experiment showed a turbulent Sc over bare ground that averaged 0.6. This is a departure from the classical assumption of one, but it agrees with other atmospheric and wind tunnel experiments that found Sc less than one. Our Sc observations also exhibited large variability. For the 19 observation periods the standard deviation was 52% of the average value, with no obvious correlation to surface atmospheric conditions. Some of this variability is likely due to measurement uncertainty, but we believe, it also reflects the actual variability in Sc.

[3] If we assume a standard neutral stability parameterization of σ_{w} (1.25 u_*) and the dissipation rate ϵ (u_*^3/kz), $Sc = 0.6$ implies a universal constant $C_0 = 4.3$ (this is discussed in more detail in Wilson et al., 2001).

The value of *Sc* has important implications for measurement of trace gas emissions. Most flux-gradient formulae assume higher values of *Sc*, which in our case leads to the underestimation of the tracer emission rate Q. It is interesting that Simpson et al. (1995) and Wagner-Riddle et al. (1996), when using FG formula similar to that investigated here, scale-up their estimates of Q by 30%. This would be consistent with *Sc* near 0.7 (they justify this adjustment as giving better agreement with energy balance observations). Dispersion modelers must also be attentive to the value of *Sc*. In using a Lagrangian stochastic model to estimate Q from concentration observations, we showed that by adjusting the dispersion model to give $Sc = 0.6$, we achieved better estimates of Q. However, the large variability in *Sc* inferred from our measurements, if true, implies that these micrometeorological methods to deduce Q for short intervals (of order 1 h) will be subject to large random error.

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